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This is the final report on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high-pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. A method is presented for vastly improving the output of the synchronously mode-locked dye laser systems. The beat frequency is increased to 155.7 kHz. Subnanosecond excited-state lifetimes for Na are obtained with only 128 averages, for the first time allowing data to be obtained within the time scale of turbulence. The first quantitative evaluation of the ASOPS technique in a flame environment is presented.

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ASYNCHRONOUS OPTICAL SAMPLING FOR LASER-BASED COMBUSTION DIAGNOSTICS IN HIGH-PRESSURE FLAMES

Final Report
Air Force Office of Scientific Research
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Abstract

This is the final report on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high-pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. A method is presented for vastly improving the output of the synchronously mode-locked dye laser systems. The beat frequency is increased to 155.7 kHz. Subnanosecond excited-state lifetimes for Na are obtained with only 128 averages, for the first time allowing data to be obtained within the time scale of turbulence. The first quantitative evaluation of the ASOPS technique in a flame environment is presented.

1. RESEARCH OBJECTIVES

The overall goal of this research is to develop and test a new combustion diagnostic for the quantitative measurement of both major and minor species in high-pressure flames. The proposed technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond laser spectroscopy which should yield a better signal-to-noise ratio (SNR) than laser fluorescence measurements in rapidly quenched combustion environments. Furthermore, ASOPS will allow determination of both quenching rates and state-to-state relaxation rates which are necessary for quantitative applications of both laser-induced and laser-saturated fluorescence at high pressure. The ASOPS technique produces a coherent signal-carrying beam and thus requires no more optical access to practical combustion devices than LDV measurements.

2. RESEARCH STATUS

2.1 Final Progress

In past experir nts, visible ASOPS studies of atomic sodium in an atmospheric-pressure flame proved to be quite successful. In order to evaluate the difficulty of using UV beams, the next subject of study was atomic indium. Although a signal was obtained, the signal-to-noise ratio (SNR) was less than expected. Experimental detection of the hydroxyl radical using ASOPS was not successful, despite our early estimates of SNR, and despite several attempts at reducing the noise emitted from the laser systems. Research during the past year has consisted, in part, of a systematic procedure to explain exactly why the above scenario was observed.

Several important improvements to the operation of the ASOPS lasers have been implemented, resulting in vastly enhanced ASOPS signals. This has allowed us to obtain a subnanosecond ASOPS decay of the 3*P* level of atomic sodium using an acquisition rate of 155.7 kHz with only 128 averages. For the first time, the ASOPS instrument has been used to take quantitative concentration measurements.

2.1.1 Improvement of Laser Performance

The dye laser contributes to noise from dye jet imperfections. New sapphire dye jets have shown a dramatic decrease in the noise bandwidth compared to specially constructed stainless-steel jets.[1] A sapphire jet nozzle has been added to each of our dye lasers. The increased stability of the dye-laser output is demonstrated in Fig. 1, which contains an FFT of the dye-laser power out to 250 kHz. The noise drops sharply until about 150 kHz. At this point, the noise floor is defined by the FFT feature of the Tektronix 602A digitizing signal analyzer. The noise actually continues to several megahertz. Nevertheless, the noise level at 150 kHz is much more favorable than at the frequency of 10 kHz where previous ASOPS measurements were taken.

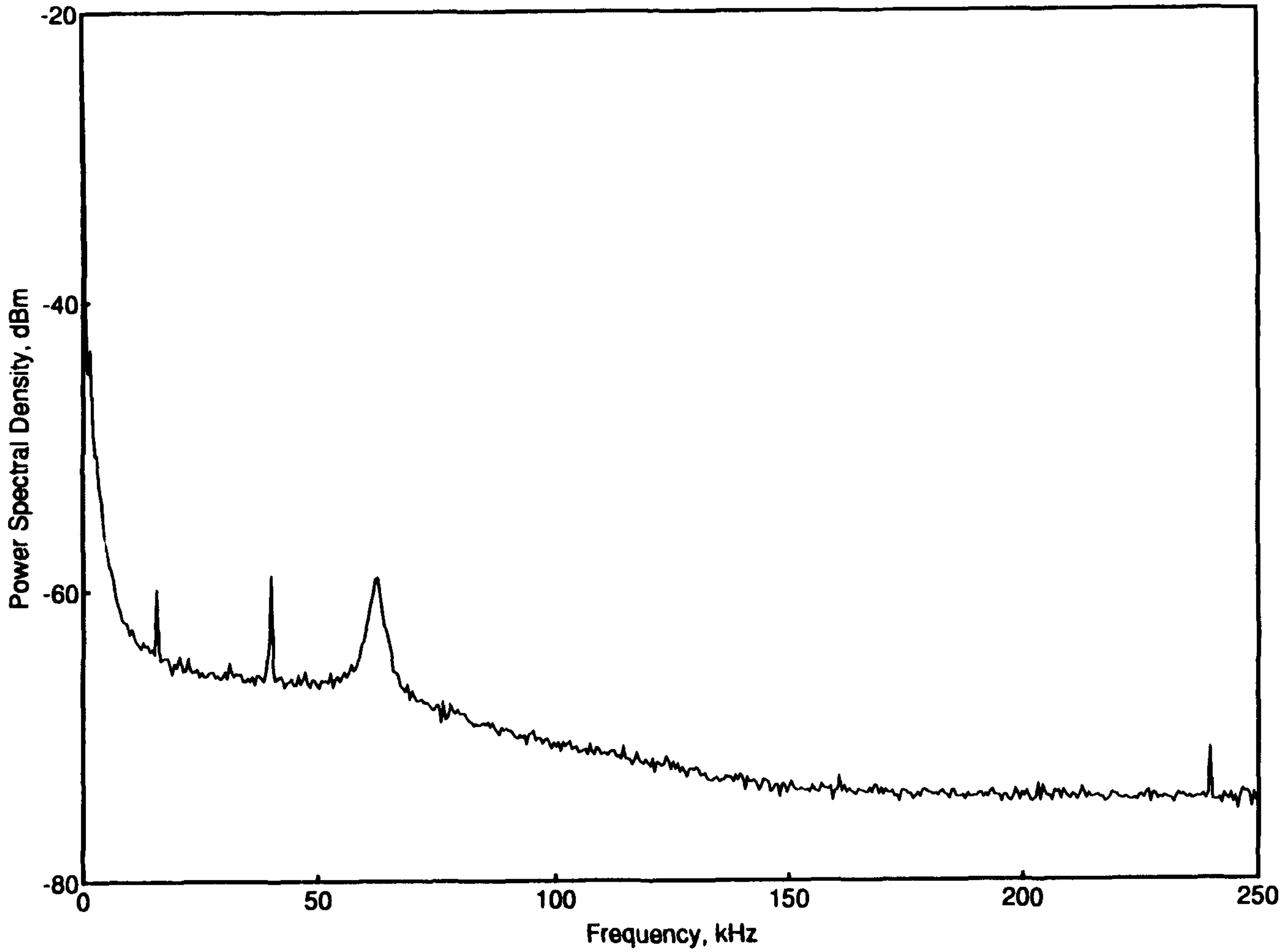

Figure 1. Dye laser noise that results when the cavity length is set at its optimum position. The curve that is shown is the result of 128 averages of individual FFT's.

This region of low noise prompted a modification of the mode-locking frequencies for each laser. The mode-locking frequency of the pump Nd:YAG laser was increased by decreasing the cavity length. The limiting value was found to be caused by the maximum mode-locking frequency that the mode-locking prism could support. The mode-locking frequency of the probe Nd:YAG laser was decreased by increasing the cavity length. The minimum value was found when the maximum setting on the translation stage was reached. Should it someday become necessary, the translation stage could be machined to allow longer cavity lengths, although no such modifications have yet been attempted. The resulting beat frequency is 155.7 kHz, which is sufficiently high to take advantage of the noise characteristics of our lasers. More importantly, this large frequency allows large numbers of averages to be taken within the time scale of turbulence.

2.1.2 Quantitative Results

ASOPS measurements are made quantitative by making simultaneous measurements using atomic absorption spectroscopy (AAS).[2] In this process, the concentration of Na is varied in the flame, and this value is measured with the AAS instrument. The ASOPS signal is then recorded on a lock-in amplifier for each concentration. The resulting calibration curve is shown in Fig. 2. The calibration curve begins to drop at high Na concentrations due to the onset of optically-thick conditions. Next, the ASOPS signal is monitored on an oscilloscope. Each signal is the result of only 128 averages. Two curves are shown in Fig. 3, with top and bottom corresponding to 1.8×10^{11} cm^{-3} and 5×10^{9} cm^{-3}, respectively. The bottom curve has a signal-to-noise ratio (SNR) of approximately 1:1. This is limited mainly by rf interference that occurs at ~2 MHz. If the source of this noise could be eliminated, the detection limit could be improved by at least an order of magnitude.

2.2 Current Research

Although the above results are impressive, substantial improvements to the basic ASOPS instrument are still being implemented. An electrooptic modulator (Quantum Technology Model 3010) has been added to the pump laser. This makes it possible to shift the ASOPS harmonics beyond the noise corner frequency, as shown in Fig. 4, where the ASOPS signal appears as sidebands about the electrooptic modulation frequency. The modulated ASOPS (MASOPS) instrument diagram is shown in Fig. 5. The two independently mode-locked laser systems of the conventional ASOPS instrument remain intact. To maintain a constant phase walk-out between the pump and probe pulse trains, the frequency synthesizers of each laser are operated in a master-slave configuration, referenced to the same 10-MHz oscillator. To ensure a constant number of pump and probe beam pulses per modulator cycle, the frequency synthesizer

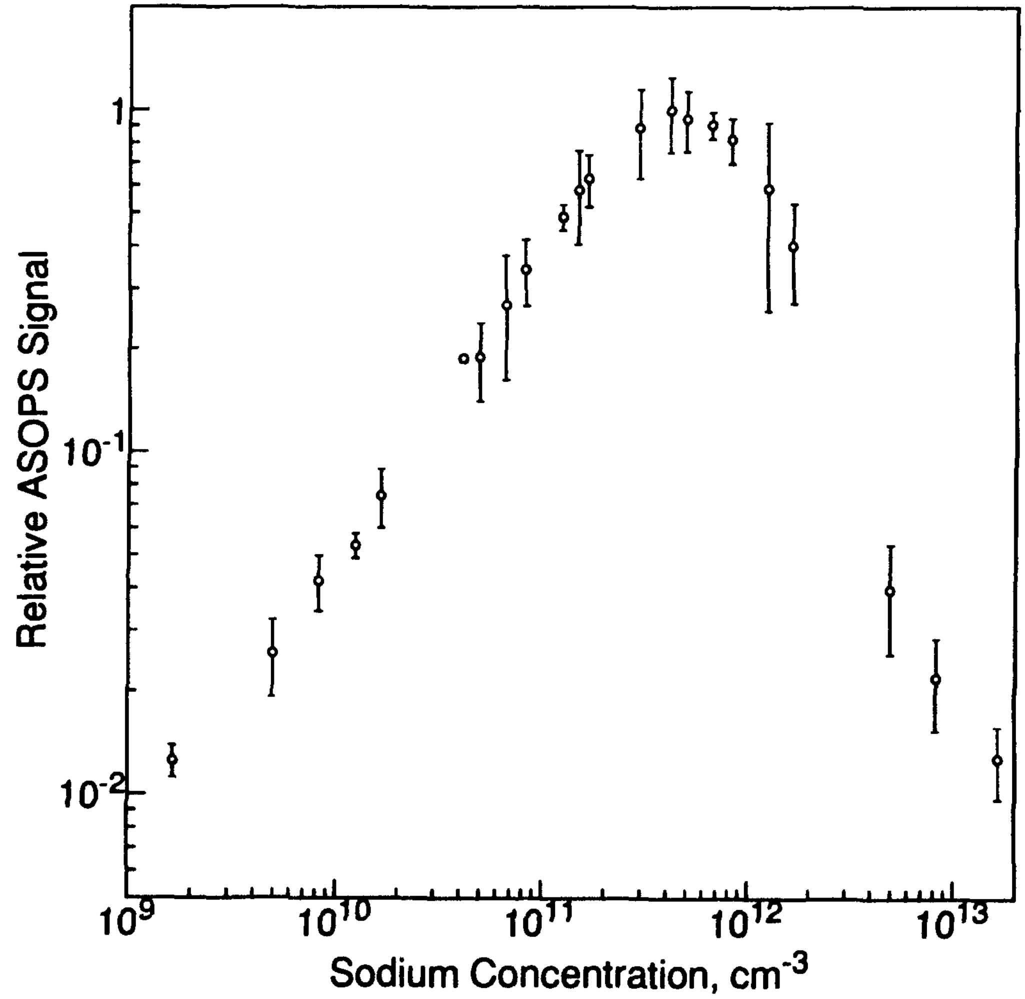

Figure 2. Variation of the ASOPS signal with sodium number density. Each point is an average of several points, ranging from three to five values. The error bars represent the sample standard deviation (1σ).

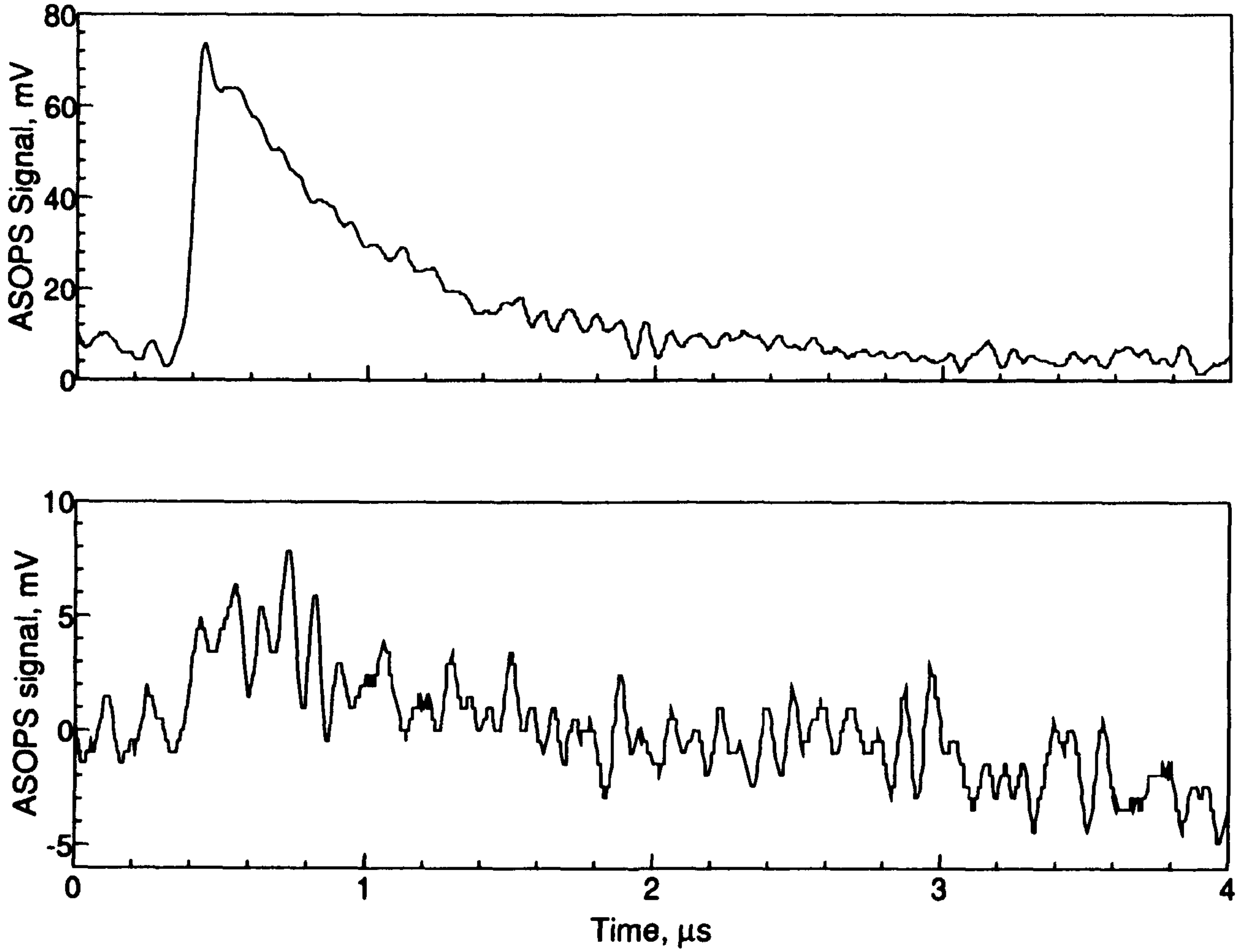

Figure 3. ASOPS decays obtained with a beat frequency of 155.7 kHz and only 128 averages. Both the pump and probe beams are set at 589.00 nm. The top and bottom plots were obtained with sodium concentrations of 1.8×10^{11} cm^{-3} and 5×10^{9} cm^{-3}, respectively.

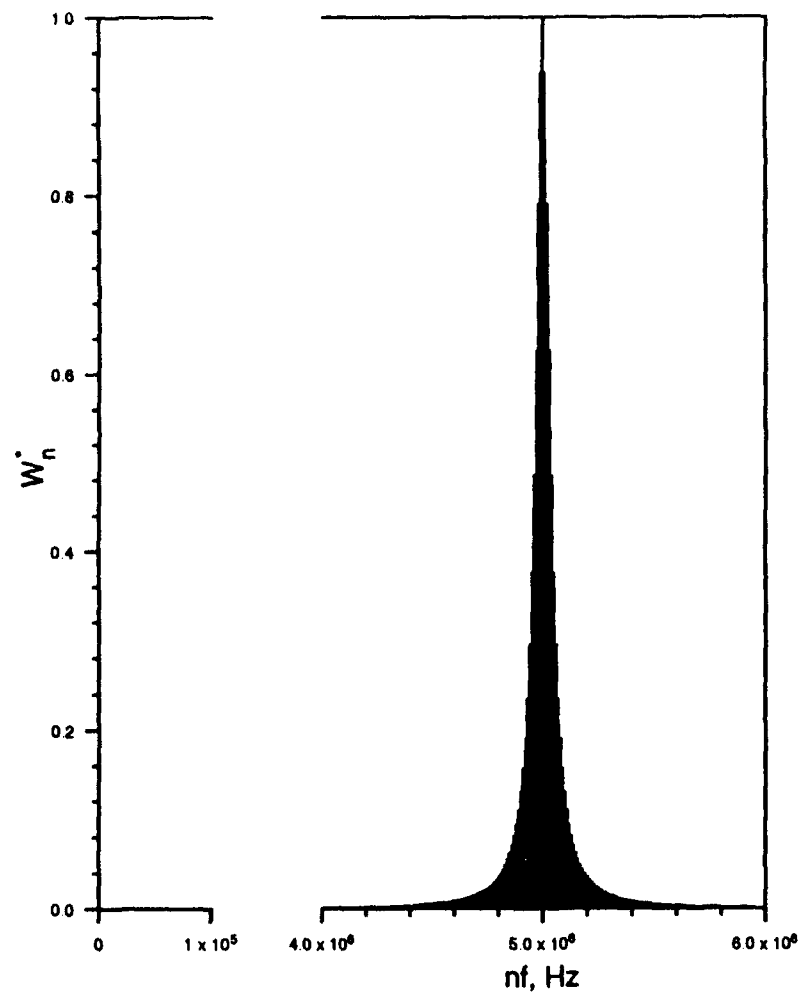

Figure 4. Spectrum that results when the ASOPS signal is obtained with a sinusoidally modulated pump beam. The modulation frequency is 5 MHz in this example.

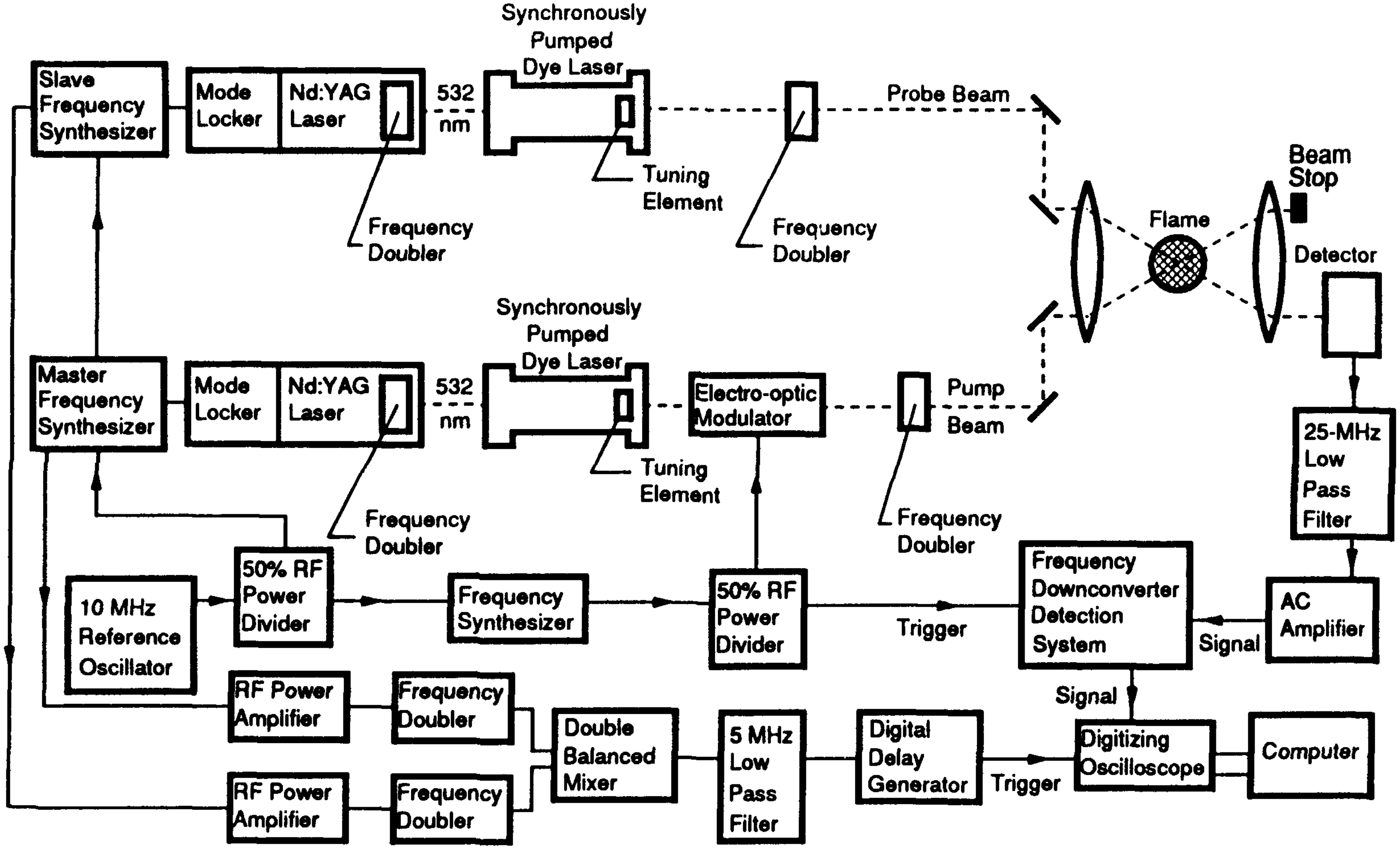

Figure 5. Modulated ASOPS (MASOPS) detection instrument.

for the modulator is also referenced to the 10-MHz oscillator. After the pump and probe lasers are crossed in the flame, the probe is again directed to a suitable detection system, and the resultant output is filtered and amplified.

The MASOPS signal must then be demodulated, so that the temporal information that is contained in the sidebands can be analyzed. This is accomplished using the frequency downconverter detection system of Fig. 6.[3] The output of a second 10-MHz oscillator is split into two portions. One portion is mixed with half of the modulator frequency-synthesizer output, which can be set at 9.95 MHz. The other half of the 10-MHz synthesizer output is mixed with the filtered and AC-amplified signal. The resulting 50-kHz signal is detected with a low-frequency lock-in amplifier (Stanford Research Systems SR510), triggered by the down-shifted modulator reference signal. The output of the lock-in amplifier is identical to the ASOPS signal of our previous instrument, and is thus directed to a digitizing oscilloscope, which is triggered at the beat frequency.

2.3 Future Work

When the frequency downconverter detection system has been constructed, the MASOPS instrument will be initially tested by again detecting atomic sodium. This will be aided by the familiarity we have with the detection of this atom. Upon completion of the visible MASOPS studies, frequency doublers will again be added to the pump and probe beams, and OH detection will be attempted.

3. PUBLICATIONS AND PRESENTATIONS

The following paper has been accepted for publication.

1. G. J. Fiechtner, G. B. King, N. M. Laurendeau, and F. E. Lytle, "Determination of Relative Number Density and Decay Rate for Atomic Sodium in an Atmospheric Premixed Flame by Asynchronous Optical Sampling", in press, *Applied Optics.*

4. RESEARCH PERSONNEL

Professors Galen B. King and Normand M. Laurendeau in the School of Mechanical Engineering and Professor Fred E. Lytle in the Department of Chemistry are co-principal investigators for this research. Mr. Gregory Fiechtner, a mechanical engineering Ph.D. candidate, joined the group in July, 1986 as a M.S. candidate, completing his M.S. thesis in August of 1989. He will receive his Ph.D. degree in May of 1992, and will leave the ASOPS project. Mr. Brian Thompson joined the group in August of 1990 as an M.S. candidate in mechanical engineering. He will receive his M.S. degree in August of 1992, and will leave the group during the summer.

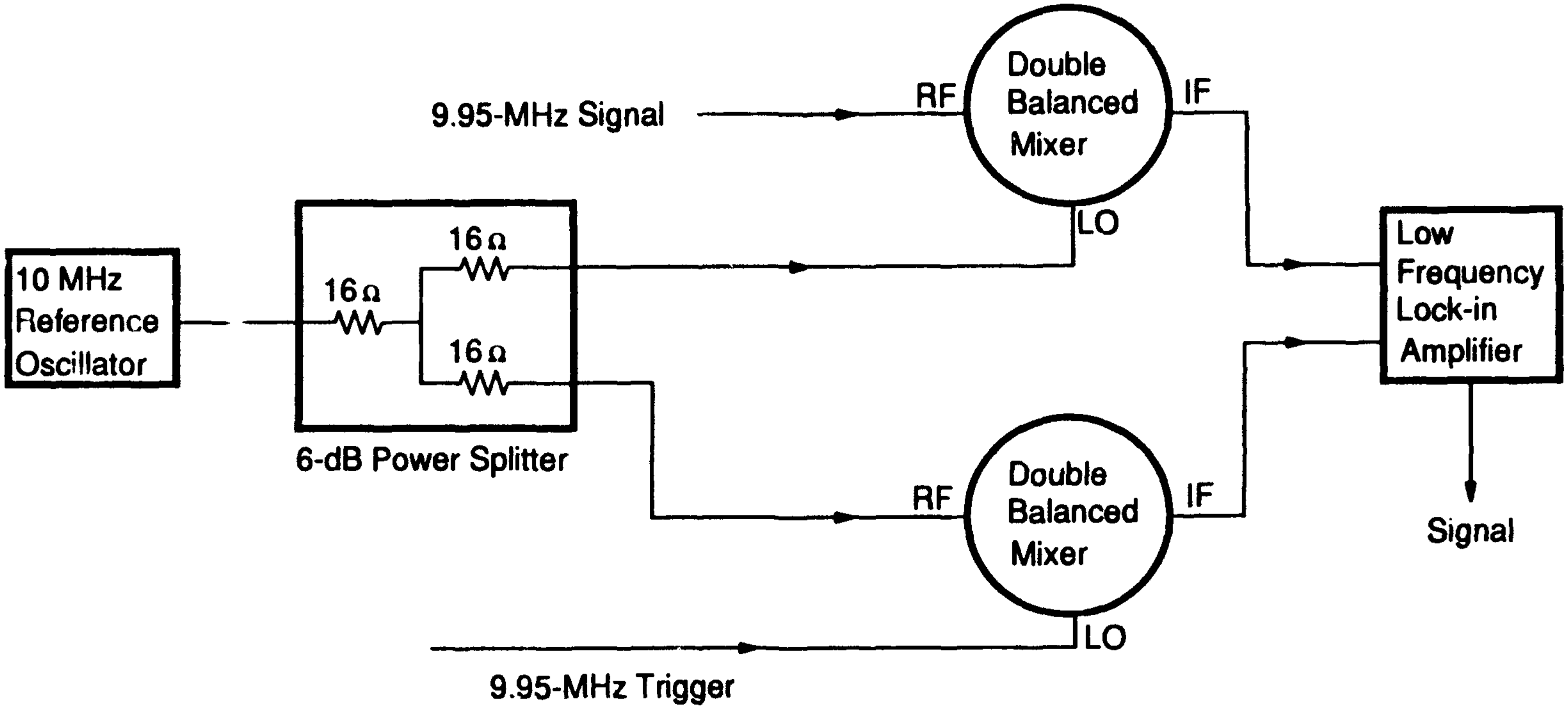

Figure 6. Frequency downconverter detection system.[3]

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2. C. Th. J. Alkemade, Tj. Hollander, W. Snelleman and P. J. Th. Zeegers, *Metal Vapours in Flames*, Pergamon Press, New York, NY (1982).

3. K. J. Weingarten, *Gallium-Arsenide Integrated Circuit Testing Using Electrooptic Sampling*, Ph.D. Dissertation, Stanford University, 1988.